

# Superconductivity at 36 K in Gadolinium-arsenide Oxides $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$

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In this paper we report the fabrication and superconducting properties of  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$ . It is found that when  $x$  is equal to 0.17,  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  is a superconductor with the onset transition temperature  $T_c^{\text{on}} \approx 36.6\text{K}$ . Resistivity anomaly near 130K was observed for all samples up to  $x = 0.17$ , such a phenomenon is similar to that of  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$ . Hall coefficient indicates that  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  is conducted by electron-like charge carriers.

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## I. INTRODUCTION

Since the discovery of superconductivity in iron-based layered quaternary compound  $\text{LaOFeP}^1$ , extensive efforts have been devoted to find new superconductors among this system.<sup>1,2,3,4,5,6,7,8,9,11</sup> It is found that with the replacement of  $P$  by  $As$  and partial substitution of  $O$  with  $F$ ,  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  changes into superconducting state below  $T_c \approx 26\text{K}$ .<sup>3</sup> Subsequently superconductivity at 25 K was also observed in  $\text{La}_{1-x}\text{Sr}_x\text{OFeAs}$  in which no  $F$  was added into the sample, therefore it was a hole-doped superconductor.<sup>4</sup> More recently, superconductors  $\text{LnO}_{1-x}\text{F}_x\text{FeAs}$  with light rare-earth substitution ( $\text{Ln}=\text{Ce}, \text{Pr}, \text{Sm}$ ) were realized and superconducting transition temperature ( $T_c$ ) was raised to 52K.<sup>5,6,7,8</sup> As to the heavy rare-earth element, however, single phase could not be easily formed and no superconducting state was observed below 2K. As to the element Gadolinium which locates near the heavy rare-earth element, experimentally a drop of resistivity was observed below 10 K but with a residual resistivity down to 2 K.<sup>9</sup> So it is worth exploring further that whether  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$  is also a superconductor with much higher  $T_c$ . In this study, we report the superconducting properties of  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$ , the onset transition temperature  $T_c^{\text{on}}$  is about 36.6 K.

## II. EXPERIMENT

Polycrystalline samples  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$  ( $x=0.12, 0.15, 0.17$ ) were synthesized by conventional solid state sintering method. The raw materials are all with high purity ( $\text{Gd}_2\text{O}_3$  99.99%,  $\text{GdF}_3$  99.99%,  $\text{Fe}$  99.95%,  $\text{As}$  99.99%,  $\text{Gd}$  99.99%). The detailed synthesis method is the same as that in the papers we reported previously.<sup>10,11</sup> The as-sintered pullet is concrete ceramic-like with dark-brown surface. X-Ray diffraction measurement was performed at room temperature using an MXP18A-HF-type diffractometer with  $\text{Cu-K}_\alpha$  radiation from  $10^\circ$  to  $80^\circ$  with a step of  $0.01^\circ$ . The magnetization measurements were carried out on a Quantum Design superconducting quantum interference device (SQUID) magnetometer. The electrical resistivity and Hall coefficient were measured by a Physical Property Measurement System (PPMS, Quantum Design) with a standard six-probe method.

Fig.1 shows the X-ray diffraction (XRD) pattern of the sam-

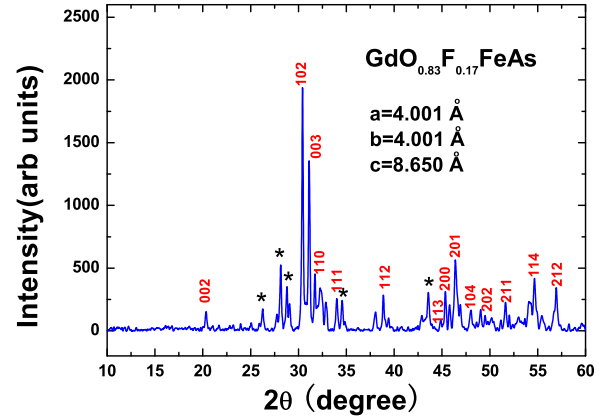


FIG. 1: (color online) XRD pattern of  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$  ( $x=0.17$ ). It is clear that the dominant phase is  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$ . The asterisks mark the peaks from the impurity phase.

ple  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$ . The pattern can be indexed in tetragonal space group with  $a = b = 4.001 \text{ \AA}$  and  $c = 8.650 \text{ \AA}$ . Obviously the phase is dominated by  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$ , though minor impurity phases still exist as marked by the asterisks. Such impurity phases could be caused by the inadequate sintering temperature  $1160^\circ \text{C}$  in our experiment. The indices of the crystal lattice we obtained are consistent with the counterparts of  $\text{LnO}_{1-x}\text{F}_x\text{FeAs}$  ( $\text{Ln}=\text{Ce}, \text{Pr}, \text{Sm}$ )<sup>5,6,7,8</sup>.

Fig.2 shows the temperature dependence of DC magnetization for sample  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$ . The diamagnetic signal appears below 22 K, a simple estimation on the magnetization at 2K reveals that the superconducting volume fraction is more than 40%. It should be noted that the response to magnetic field in the normal state of  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  is paramagnetic with a minor value compared to diamagnetic value and such a contribution to magnetization is subtracted as a background (the same to ZFC and FC curve).

The temperature dependence of resistivity of all three samples are shown in Fig.3. We can see that the resistivity shows an anomaly around 130 K, and this anomaly weakens with more  $F$  doping, such an anomaly and corresponding evolution with  $F$  doping have been observed in other Fe-based Ar-

### III. CONCLUDING REMARKS

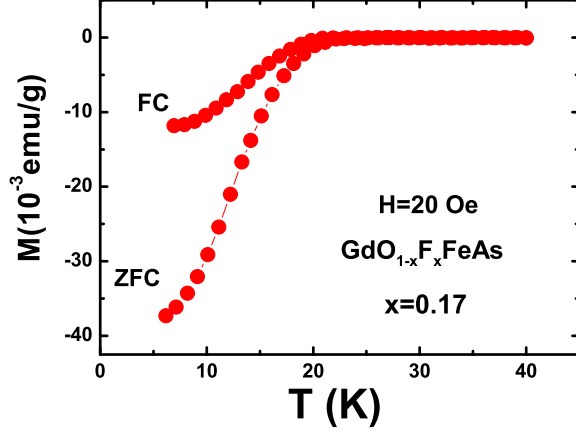


FIG. 2: (color online) DC magnetization of  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  measured in the zero-field-cooled (ZFC) and field-cooled (FC) processes. A diamagnetic signal is easily observed at about 22 K

senic compounds, however, the anomaly did not happen in Nickel based Arsenic system. The obvious disparity between Fe-based and Ni-based systems deserves to be further studied. Both  $x = 0.15$  and  $x=0.17$  samples exhibit superconducting transitions and zero-resistance at a lower temperature. From the inset of Fig.3 we can see the onset drop of resistivity about 36.6 K for  $x = 0.17$  sample. As to  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$ , a slight hump near 130 K is also observed in resistivity curve, it suggests that  $T_c$  could be increased further as long as more Fluorine were doped into  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$ . The transition width can also be narrowed in a refined fabrication process in the future.

Hall effect measurement for sample  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  was shown in Fig.4. The transverse resistivities  $\rho_{xy}$  above  $T_c$  are all negative, indicating that the normal state conduction of  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  is dominated by the electron-like charge carriers. The Hall coefficient  $R_H = \rho_{xy}/H$  changes slightly at high temperatures but drops below 100 K. The value of  $R_H$  is about  $-1 \times 10^{-8} \text{ m}^3/\text{C}$  at 100 K, compared with that of  $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$ , the value of Hall coefficient is similar.<sup>10</sup> An estimation based on the single band model gives a charge carrier density of  $1 \times 10^{21}/\text{cm}^3$ .

In this study we report the fabrication and the superconducting properties of  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$ , as  $x$  is equal to 0.17,  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  is a superconductor with the onset transition temperature of about 36.6 K. Resistivity anomaly near 130 K was observed for all samples, which is similar to that of  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$ . Hall coefficient suggests that  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$  is conducted by electron-like charge carriers.

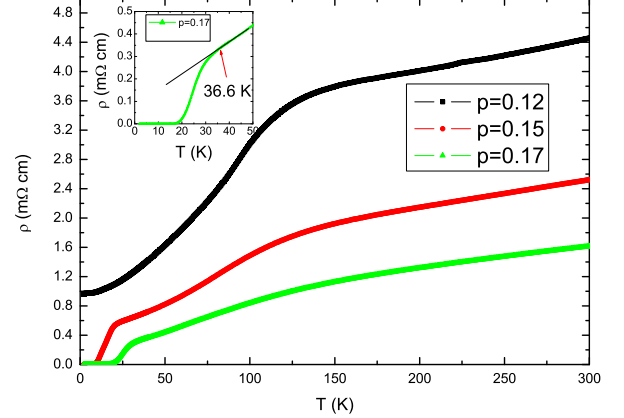


FIG. 3: (color online) The electrical resistivity vs temperature for  $\text{GdO}_{1-x}\text{F}_x\text{FeAs}$  ( $x=0.12, 0.15, 0.17$ ). The inset shows the enlarged view of superconducting transition area for the sample  $x = 0.17$ . The onset transition temperature is defined at the point where the resistivity starts to deviate from the normal state background as marked by the straight line here.

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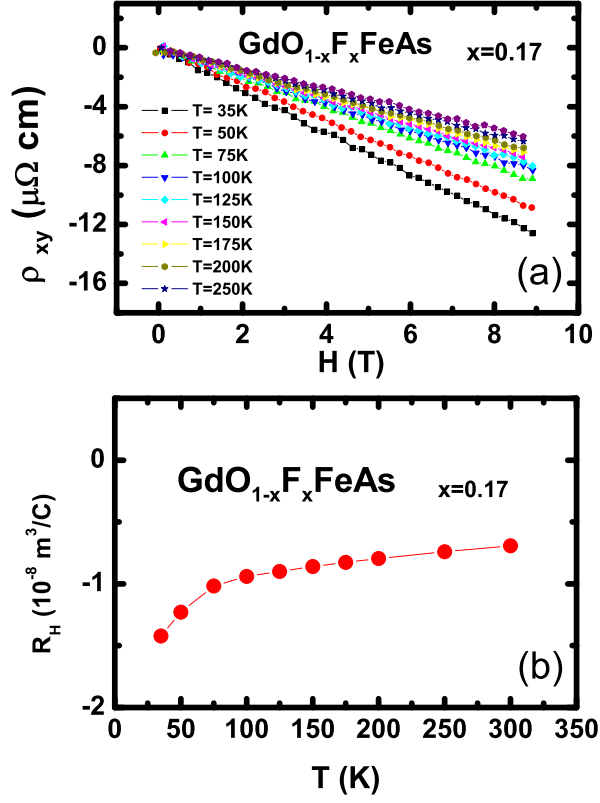


FIG. 4: (color online) (a) Transverse resistivity with relation of magnetic field at different temperatures for  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$ ; (b) Temperature dependence of Hall coefficient for  $\text{GdO}_{0.83}\text{F}_{0.17}\text{FeAs}$ , the negative value indicates that the charge carrier is electron type.

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